Muon spin rotation study of the magnetic penetration depth in the intercalated graphite superconductor CaC$_6$

D. Di Castro,[1] A. Kanigel,[2,3] A. Maisuradze,[4] A. Keren,[2] P. Postorino,[5] D. Rosenmann,[6]
U. Welp,[7] G. Karapetrov,[7] H. Claus,[7] D.G. Hinks,[7] A. Amato,[4] and J. C. Campuzano[3,7]

[1]*CNR-SPIN and Dipartimento di Ingegneria Meccanica,*
Università di Roma Tor Vergata, Via del Politecnico 1, I-00133 Roma, Italy
[2]*Department of Physics, Technion, Haifa 32000, Israel*
[3]*Department of Physics, University of Illinois at Chicago, Chicago, IL 60607*
[4]*Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland*
[5]*Dipartimento di Fisica, Università di Roma La Sapienza, Piazzale Aldo Moro 2, I-00185 Roma, Italy*
[6]*Center for Nanoscale Materials, Argonne National Laboratory, Argonne, IL 60439*
[7]*Materials Science Division, Argonne National Laboratory, Argonne, IL 60439*
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We report temperature- and magnetic field-dependent bulk muon spin rotation measurements in a c-axis oriented superconductor CaC$_6$ in the mixed state. Using both a simple second moment analysis and the more precise analytical Ginzburg-Landau model, we obtained a field independent in-plane magnetic penetration depth $\lambda_{ab}(0) = 72(3)$ nm. The temperature dependencies of the normalized muon spin relaxation rate and of the normalized superfluid density result to be identical, and both are well represented by the clean limit BCS model with $2\Delta/k_BT_c = 3.6(1)$, suggesting that CaC$_6$ is a fully gapped BCS superconductor in the clean limit regime.

Recently the field of graphite intercalated compounds (GICs) has regained attention after the discovery of the superconducting GIC CaC$_6$ with T$_c$ ~11.5K.[1,2] Soon after the discovery it was suggested[3] that CaC$_6$ is an unconventional superconductor, and that the pairing interaction is of electronic origin. As time elapsed, both calculations[4] and experiments showed that CaC$_6$ is a conventional BCS superconductor. In particular, experiments revealed the existence of a significant Ca isotope effect,[5] the absence of gap nodes,[6] a single gap[7] with BCS temperature dependence,[8] as well as the BCS temperature dependence of the London penetration depth .[9] Moreover, calculations,[4] supported by experimental evidence ,[6,7,10–13] showed the importance of a Ca derived interlayer band, crossing the Fermi energy, which have sufficiently strong coupling with both in-plane intercalant and out-of-plane graphite phonon modes to explain the T$_c$ of CaC$_6$ within a standard electron-phonon coupling mechanism. On the other hand, the coupling between graphene-derived electrons and high-frequency graphene-derived phonons was also demonstrated to be relevant.[14]

At present the mechanism of superconductivity in CaC$_6$ and most of its properties have been investigated and clarified. Nevertheless a complete description of the superconducting state in a Type II superconductor requires a whole coherent set of parameters measured with high precision, such as the value of the coherence length ξ and the magnetic penetration depth λ. These points are somehow still lacking. Indeed, two very different values of the in plane penetration depth lambda $\lambda_{ab}(0)$ have been reported in the literature[9,15] and the temperature dependent superfluid density, $\rho(T)$, has been measured with surface sensitive technique only.[9] For the extremely air sensitive superconductor CaC$_6$, bulk measurement of

the superconducting properties would be thus very important. We also notice that a new unambiguous bulk determination of $\rho(T)$ could also help in clarifying the nature of the CaC$_6$ conduction regime, since both dirty limit[9,16,17] and clean limit[15,18] regime have been reported so far. This system is also interesting from the point of view of μSR data analysis, since it is a low $\kappa = \lambda/\xi$ superconductor with reduced, in field, vortex phase.

The muon spin rotation (μSR) technique is known to be one of the most indicated techniques to measure the bulk properties of type II superconductors in the vortex state. In this paper we report a transverse field muon spin rotation (TF-μSR) experiment on high quality CaC$_6$ c-axis oriented samples, aimed to probe the vortex lattice and determine $\xi_{ab}(0)$, $\lambda_{ab}(0)$, and the temperature dependence of the superfluid density $\rho(T)$.[19] By using a peculiar method developed to collect reliable μSR data on very thin samples, such as the CaC$_6$ ones, we obtained $\lambda_{ab}(0) = 72(3)$ nm and $\xi_{ab}(0) \simeq 38$ nm. The penetration depth results to be field independent, suggesting a fully gapped Fermi surface. The temperature dependence of the normalized superfluid density is very similar to that one of the normalized muon spin relaxation rate, as it is expected for a $\kappa \simeq 2$ superconductor at fields about $B_{c2}/2$. Both the temperature dependencies are well represented by the clean limit BCS model, with $2\Delta/k_BT_c = 3.6(1)$, suggesting that CaC$_6$ is a BCS superconductor in the clean limit regime.

In TF-μSR the sample is cooled-down in a magnetic field, which here was parallel to the c-axis. The muons are implanted in the sample with their initial spin polarization almost perpendicular to the external magnetic field. Each muon precesses around to the local field. When the superconducting sample is in the mixed state,

each muon probes a slightly different field. As a result, a de-phasing process takes place and the average muon spin polarization decays. From the average polarization of the muon as a function of time, the parameters of the vortex lattice, λ and ξ, can be extracted.[20,21]

CaC_6 samples were prepared using the alloy method as described by Emery *et al.*.[2] A stainless steel tube is loaded with Lithium and Calcium in the ratio 3:1. Natural graphite flakes are then added to the ampoule. The reaction takes place in an argon atmosphere for 10 days at 350 C. The flakes are typically of a size 2 mm × 2 mm, with a thickness of 50 μm. The low-field superconducting transition onset is at $T_c = 11.6$ K.[22] It was reported previously that these samples are very reactive and tend to degrade very fast. This is observed as an increase in the transition width and a decrease in the superconducting volume fraction. For that reason the samples were kept in an He atmosphere and overall were exposed to the room atmosphere for not more than a few minutes.

The experiments were done in the GPS spectrometer at the Paul Scherrer Institute PSI Villigen, Switzerland. Three flakes were used, covering together an area of about 4 mm × 4 mm. The CaC_6 samples are too thin to stop the muons. Given the muon stopping distance and the density of CaC_6, a sample of a few hundred microns is needed for this purpose. To maximize the number of muons stopping in the samples, we used a method developed to measure very thin crystals.[23] The samples were sandwiched between two stacks of Kapton foil rectangles. This assembly is illustrated in the panel (a) of Fig. 1. Each Kapton foil is 125 μm thick. The role of the Kapton foils in front is to slow down the muons so that most of them will stop in the sample. The typical statistics of our μSR measurements was about 17 millions events.

Muons in Kapton are known to have a very high probability of forming muonium (muon-electron bound state) However, a muonium signal is not observed in our experiment. In addition, muons stopped by Kapton do not contribute substantially to the measured asymmetry since it is known that the asymmetry is strongly reduced by a thick Kapton layer.[23] The Kapton foils in the back of the sample prevent slow muons that did not stop in the sample from stopping in the windows of the cryostat. In addition, we used a veto detector, that reduces the background signal by removing events from the data where the incoming muon missed the sample. Using the veto counter and Kapton combination we were able to get an asymmetry of about 0.1, about half of that we estimated to be "good" signal coming from the CaC_6 sample.

The μSR signal was recorded in the usual time-differential way by counting positrons from decaying muons as a function of time. The time dependence of the positron rate is given by the expression :[24]

$$N(t) = N_0 \frac{1}{\tau_\mu} e^{-\frac{t}{\tau_\mu}} [1 + aP(t)] + N_{bg} \qquad (1)$$

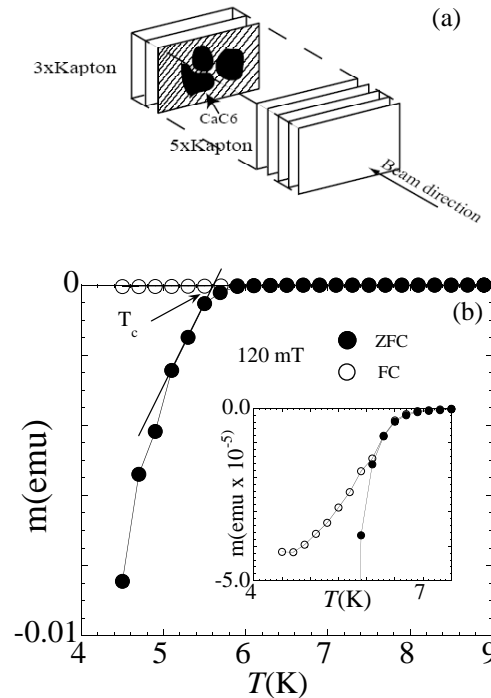where N_0 is the normalization constant, N_{bg} denotes the

FIG. 1: (a) Illustration of the CaC_6 sample and the Kapton stacks used to degrade the muon beam (see text). (b) Magnetic moment of CaC_6 as a function of temperature measured at the same field used in TF-μSR measurements (120 mT). The closed (open) symbols represent zero-field cooling (field cooling) measurements. Inset to panel (b): magnetic moment on enlarged scale to better show the ZFC data.

time-independent background, τ_μ=2.19703(4)x10^{-6} s is the muon lifetime, a is the maximum decay asymmetry for the particular detector telescope and $P(t)$ is the polarization of the muon ensemble $P(t) = \int P(B)\cos(\gamma_\mu Bt + \phi)dB$. Here $\gamma_\mu = 2\pi$ x 135.5342 MHz/T is the muon gyromagnetic ratio and ϕ is the angle between the initial muon polarization and the effective symmetry axis of a positron detector. $P(t)$ can be linked to the internal field distribution $P(B)$ by using the algorithm of Fourier transform.[24]

In Fig. 2 the magnetic field distribution $P(B)$ at an applied field of 120 mT above (7 K) and below (3 K) T_c, obtained from the measured μSR time spectra by performing fast Fourier Transform, is shown. In the normal state, $P(B)$ is a sharp symmetric line centered at the position of the external magnetic field. Below T_c the field distribution results to be composed by two signals: one is represented by a sharp symmetric peak centered at the position of the external field and ascribed to different sources of background (cryostat window, Kapton foils), and the other is a broadened line, which signals the formation of a vortex lattice (VL) in the supercon-
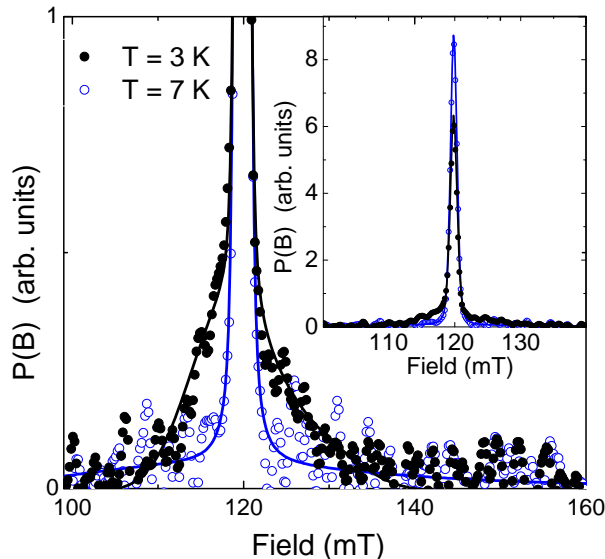
FIG. 2: Magnetic field distribution $P(B)$ at an applied field of 120 mT above (7 K) and below (3 K) T_c obtained from the measured μSR time spectra by means of fast Fourier Transform. The solid lines are fit by using Eq. 6. Inset: the same as in the main panel on larger scales.

ducting part of the sample. Indeed, VL makes the field in the sample highly inhomogeneous. This induces an increase of the muon spin relaxation rate σ, which is strictly related to the second moment of the field distribution. Moreover, the line shape of the superconducting $P(B)$ is asymmetric, as expected for a field distribution within a reasonably well arranged VL. This could be questionable, since recently, it has been shown that the irreversibility line in CaC$_6$ coincides with the $H_{c2}(T)$ line,[25] which suggests that the pinning in this sample is very strong and that a large number of pinning sites are available. In the panel (b) of Fig. 1 the magnetic moment of CaC$_6$, measured at the same field used in TF-μSR measurements, is reported as a function of temperature in zero field cooling (ZFC) and field cooling (FC) conditions. In the FC conditions the magnetic moment is negligible compared to the ZFC one, indicating a lack of field expulsion. The same was observed in the μSR experiment: we found almost no change in the average muon precession frequency when entering the superconducting phase. This is typically an effect of strong pinning, which confirms previous results.[25] The regularity of the VL could be affected by the presence of pinning in the sample. The degree of the pinning induced disorder of the flux line lattice (FLL) depends on many factors, as the strength and the nature of the pinning and the value of the external field. However, the asymmetric line-shape of the measured field distribution in our samples indicates that, although a disorder of the FLL is present, it is not large enough to prevent a reliable analysis of the data.

As a first step, the μSR time spectra collected below T_c were fitted by two Gaussian lines :[26]

$$P(t) = A_{bg} \exp(-\sigma_{bg}^2 t^2/2) \cos(\gamma_\mu B_{bg} t + \phi) + \quad (2)$$
$$A \exp(-\sigma^2 t^2/2) \cos(\gamma_\mu B t + \phi)$$

where $A_{bg}, \sigma_{bg}, B_{bg}$ and A, σ, B are the asymmetry, the relaxation rate and the mean field of the background signal and of the superconducting signal, respectively. The first one is assumed to be temperature- and field-independent. The second one is an approximation to the field distribution in the superconducting state of the sample. This distribution is both temperature- and field-dependent, and it generates a Gaussian relaxation rate $\sigma(T)$ of the muon spin polarization, which is proportional to the second moment of the local field distribution. The minimum field for which we could obtain a reliable value for σ is 60 mT. Below this field the combination of a very fast relaxation and a relatively low frequency prevents a good fit.

In Fig. 3 we show the field dependence of σ measured at $T \simeq 2$ K. The measurements were done in field-cooled conditions: for every field value the sample was warmed to a temperature above T_c and cooled back to about 2 K. As can be seen, there is an almost linear decrease in the relaxation rate with increasing field.

For a fully gapped BCS superconductor, as CaC$_6$ is supposed to be,[4,6,7,9] the magnetic penetration depth λ should be field independent.[27,28] In this case, by the analysis of the field dependence of σ it is possible to obtain an estimate of the absolute value of λ, by using the approximation developed by Brandt [Eq.(13) in Ref.29], which is considered a very good one for superconductors with $\kappa = \lambda/\xi \geq 5$ (ξ is the coherence length):

$$\sigma \approx 0.172 \gamma_\mu \frac{\Phi_0}{2\pi} \left(1 - \frac{B}{B_{c2}}\right) \left[1 + 1.21 \left(1 - \sqrt{\frac{B}{B_{c2}}}\right)^3\right] \lambda_{ab}^{-2}$$
$$(3)$$

where $\gamma_\mu = 2\pi \times 135.5342$ MHz/T is the muon gyromagnetic ratio, Φ_0 is the flux quantum, B_{c2}^{ab} the in plane upper critical field, and λ_{ab} the in plane magnetic field penetration depth. The model reproduces the data reasonably well (solid line in Fig. 3). This is thus consistent with a field independent λ_{ab} and, therefore, with CaC$_6$ being a fully gapped superconductor. The fit (done without considering the data point at 250 mT which is too close or above B_{c2} and is compatible with the background value) gives $\lambda_{ab} = 77(3)$ nm and $B_{c2} = 230(10)$mT. The latter, within the Ginzburg-Landau picture, corresponds to $\xi_{ab} \simeq 38$ nm. Both the obtained parameters, ξ_{ab} and λ_{ab}, are in good agreement with previous results[2,6,7,9,10,15,30] and give $\kappa = \lambda_{ab}/\xi_{ab} \approx 2$. Although the $\kappa = \lambda/\xi = 2$ is rather small the Eq. 3 is still valid in the fields $\sqrt{B/B_{c2}} \geq 0.5$ (see Fig. 6 in Ref.29), that is, in the range of our data.
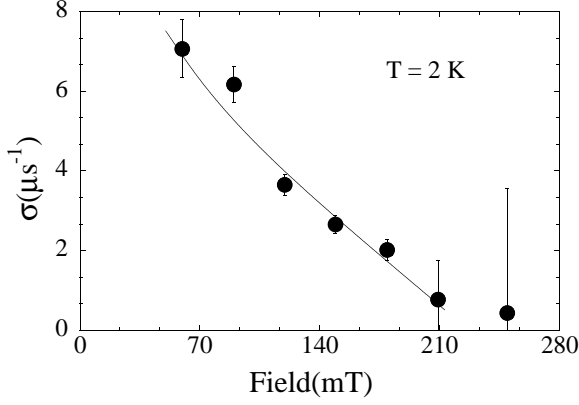
FIG. 3: Gaussian relaxation as a function of magnetic field at T = 2 K. The solid line is a fit by Eq. 3

These results have been obtained by using a gaussian approximation for the field distribution $P(B)$. To obtain a more accurate value of λ_{ab} in the following we will analyze the muon data by using the analytical Ginzburg-Landau (AGL) approximation ,[31,32] which is valid for a broad range of fields and of Ginzburg-Landau parameter κ.

The spatial field distribution $\mathbf{B(r)}$ in the vortex lattice was calculated as follows:

$$B(\mathbf{r}) = \langle B \rangle \sum_{\mathbf{G}} \exp(-i\mathbf{G} \cdot \mathbf{r}) B_{\mathbf{G}}(\lambda, \xi, \langle B \rangle). \quad (4)$$

The $B_{\mathbf{G}}$ are the Fourier components:[31,32]

$$B_{\mathbf{G}} = \frac{\Phi_0}{S} \frac{f_\infty K_1[\frac{\xi_v}{\lambda}(f_\infty^2 + \lambda^2 G^2)^{1/2}]}{(f_\infty^2 + \lambda^2 G^2)^{1/2} K_1(\frac{\xi_v}{\lambda} f_\infty)}, \quad (5)$$

with $f_\infty = 1 - b^4$, and

$$\xi_v = \xi(\sqrt{2} - \frac{0.75}{\kappa})(1 + b^4)^{1/2}[1 - 2b(1 - b)^2]^{1/2}.$$

Here, $\langle B \rangle$ is the average magnetic field inside the superconductor, ξ the coherence length, $K_1(x)$ is the modified Bessel function, $\mathbf{r}$ the vector coordinate in a plane perpendicular to the applied field, $\mathbf{G} = 4\pi/\sqrt{3}a(m\sqrt{3}/2, n+ m/2)$ the reciprocal lattice vector of the hexagonal FLL lattice, a the intervortex distance, and m and n are integer numbers. From this spatial field distribution the probability field distribution $P(B)$ was calculated.

The μSR time spectra were fitted with the depolarization function P(t) obtained from P(B) as follows:

$$P(t) = A \cdot e^{-1/2\sigma_g^2 t^2 + i\phi} \int P(B)e^{i\gamma_\mu Bt} dB + \quad (6)$$
$$A_{bg} \cdot e^{-\sigma_L t + i(\gamma_\mu B_a t + \phi)},$$

where the second term is the signal of the background with asymmetry A_{BG} and relaxation σ_L. The parameter

σ_g describes the disorder of the FLL and was fixed proportional to $1/\lambda^2$ such that $\sigma_g(0) = 2.1$ μs^{-1} (note that this relaxation is much smaller than the superconducting relaxation, which are summed in quadrature). Such relation corresponds to the rigid (well pinned) FLL.[21,33] We neglected additional nuclear relaxation, since the concentration of Ca and C isotopes with magnetic nuclear moments is practically zero. The parameter ξ was kept fixed during the fit at the values determined, at each temperature, from the corresponding $B_{c2}(T)$ via the Ginzburg-Landau relation $\xi(T) = \sqrt{(\Phi_0/(2\pi B_{c2}(T)))}$. The $B_{c2}(T)$ values were obtained by linearly interpolating the following four experimental points: $B_{c2}(2K) = 230$ mT, given above by the fit of the field dependence of σ, and $B_{c2}(5.6K) = 120$ mT, $B_{c2}(8.5K) = 60$ mT, $B_{c2}(11.5K) = 0$ measured from magnetization experiments (only the measurement at 120 mT is shown in Fig. 1). These experimental points agree well with the $B_{c2}(T)$ curve reported by Cubitt et al..[15] This model represents very accurately the experimental data, as shown by the solid line in Fig. 2.

In Fig. 4, the values of λ_{ab}, obtained by fitting the μSR data with Eq. 6 at the lowest temperature and several fields, are shown. No field dependence of λ_{ab} is detected within error-bar, as expected for conventional fully gapped BCS superconductors and already suggested by the second moment analysis (Fig. 3). A weighted average of the data gives $\lambda_{ab} = 72(3)$ nm, compatible, within the errors, with the value obtained by fitting the field dependence of the relaxation rate, and in very good agreement with the value reported in Ref. 9. Recently Cubitt et al.[15] reported a magnetic penetration depth $\lambda = 50$ nm using SANS on CaC_6 compound. In their analysis they used the London model with Gaussian cut off (LGC). As it was shown previously by Yaouanc et al.,[32] the form factors of the LGC model deviate substantially from the exact solution of the Ginzburg-Landau model in a very broad range of fields. Indeed, an analysis of our data by using the LGC model gives $\lambda_{ab} = 52(4)$ nm, in good agreement with the results of Cubitt et al.[15] The AGL model used here gives more reliable results ,[32] and systematic deviations of the AGL model from the exact solution of the Ginzburg-Landau[34] model are within the error bar.

The magnetic penetration depth is defined, in the London limit of low fields, as a measure of the superfluid density $\rho \propto \lambda^{-2}$. However, for fields close to B_{c2} the mean magnitude of the Ginzburg-Landau order parameter $\psi(r)$ drops due to a substantial overlapping of the vortex cores.[34] The spatial average of the superfluid density is thus well described by:[35,36]

$$\rho(T) \propto \langle | \psi(r) |^2 \rangle \lambda^{-2}(T) \simeq (1 - B/B_{c2}(T))\lambda^{-2}(T), \quad (7)$$

where $\langle \cdots \rangle$ refers to the spatial average over the unit cell of the FLL. The temperature dependence of λ, needed to calculate $\rho(T)$ with Eq.7, was obtained by fitting of Eq. 6 to the data collected at different temperatures in
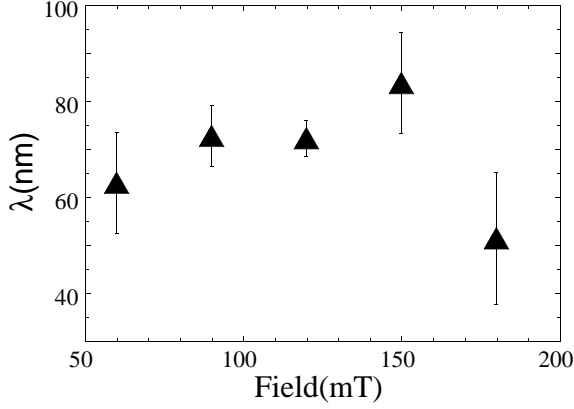
FIG. 4: Magnetic penetration depth as a function of magnetic field obtained from the fit to the μSR time spectra at 2 K.
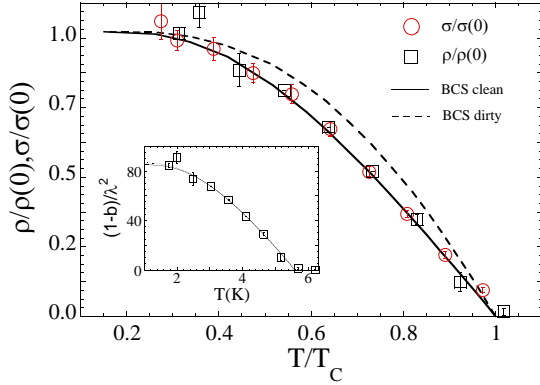

FIG. 5: Normalized superfluid density $\rho(T)/\rho(0)$ and muon spin relaxation rate $\sigma(T)/\sigma(0)$ as a function of the reduced temperature T/T_c measured at 120 mT. Full and dashed lines are BCS clean limit and dirty limit curves, respectively. Inset: temperature dependence of ρ and BCS fits.

a field of 120 mT. The $B_{c2}(T)$ values were obtained as described above.

In Fig. 5, we show the normalized superfluid density $\rho(T)/\rho(0)$ as a function of the reduced temperature T/T_c measured at 120 mT. $\rho(0)$ is obtained from the BCS fit to the temperature dependent $\rho(T)$ by using the equation:[37]

$$\rho(T) = \rho(0)[1-\frac{2}{k_BT}\int_0^\infty f(\varepsilon,T,\Delta(T))[1-f(\varepsilon,T,\Delta(T))]d\varepsilon]$$

(8)

where $f(\varepsilon,T,\Delta(T)) = [1+\exp[\sqrt{\varepsilon^2 + \Delta^2(T)}/(k_BT_C)]]^{-1}$ is the Fermi distribution and $\Delta(T) = \Delta(0)\delta(T/T_c)$, with $\delta(T/T_c)$ given by the conventional BCS temperature dependence of the gap.[38] Indeed, although data at temperatures lower than 1.75 K would be important to clearly assess the occurrence of a saturation, however the ab-

sence of a field dependence of λ points to nodeless gap in the ab-plane of CaC_6, which is consistent with previous theoretical end experimental studies.[4,6,7,9] Therefore it is correct to fit the experimental data using Eq. 8. In order to reduce the number of fitting parameters, the fit has been performed by keeping T_c fixed at the value 5.6(1) K obtained from magnetization data at the same field of 120 mT (see Fig. 1). As shown in the inset to Fig. 5, the fitting curve (solid line) represents well the experimental data, yielding $\rho(0) = 85.1(3)$ μm^{-2} and $\Delta(0) = 0.868(5)$ meV, which gives $2\Delta(0)/k_BT_c = 3.6(1)$. This value is compatible with the one expected for a weak coupling BCS superconductor and is in good agreement with previous results.[6,7,9] A fit with the dirty limit BCS temperature dependence[37] is also acceptable (see inset to Fig. 5), although with larger χ^2 and larger errors of the fitting parameters ($\rho(0) = 87(2)$ μm^{-2}, $\Delta(0) = 0.66(2)$ meV). With these parameters we would obtain $2\Delta/k_BT_c = 2.74(1)$. This quantity is substantially different from the previously reported ones[6,7,9] and it is well below the BCS weak-coupling ratio. The normalized superfluid density, shown in the main panel of Fig. 5, clearly follows the clean limit curve, and is not compatible with the dirty limit one, suggesting that bulk CaC_6 is a BCS superconductor in the clean limit regime, in agreement with Mialitsin et $al.$[18] and Cubitt et $al.$[15]

In the main panel of Fig. 5 the temperature dependence of the normalized relaxation rate $\sigma(T)$, obtained by the two gaussian model (Eq. 2), is also reported. These data closely resemble the $\rho(T)$ curve. Indeed, as follows from the Fig. 6 of Ref. 29 the quantity $\sigma\kappa^2/B_{c2}(1-b) \propto \sigma/[(1-b)/\lambda^2]$ is practically independent of the reduced field $b = \langle B\rangle/B_{c2}$ for $\kappa \simeq 2$ and $\sqrt{b} > 0.25$. On the other hand, $\sigma/[(1-b)/\lambda^2]$ is proportional to σ/ρ. Therefore, the present case with $\kappa \simeq 2$ is exceptional, when a simple second moment analysis gives a quite precise result for the temperature dependence of the superfluid density in a broad range of fields.

To summarize, we performed TF-μSR measurements on the intercalated graphite superconductor CaC_6 in the vortex-lattice state. Our experimental method included the use of Kapton tape to gather good data from very thin samples, 50 μm thick. Despite of strong pinning, we detected an asymmetric field distribution, typically observed in superconducting single crystals in the vortex state. The data were analyzed with two different models. Using a simple two-Gaussian model, the relaxation rate σ was obtained. Its field dependence suggested a field independent $\lambda_{ab}(0) = 77(3)$ nm and a coherence length $\xi_{ab}(0) \simeq 38$ nm. The more precise analytical Ginzburg-Landau model yielded $\lambda_{ab}(0) = 72(3)$ nm, a value remarkably close to that one obtained by the second moment analysis and in good agreement with previous microwave measurements.[9] Also this analysis reveal that $\lambda_{ab}(0)$ does not depend on magnetic field, suggesting a fully gapped superconductivity in CaC_6, in agreement with previous reports.[6,7,9] The temperature dependence of the superfluid density was analyzed by both the clean

and the dirty limit BCS models. The experimental data are well fitted by the clean limit one, giving a gap-to-T_c ratio $2\Delta(0)/k_B T_c = 3.6(1)$, in agreement with previous results obtained by surface sensitive techniques.[7,9] The temperature dependence of the normalized superfluid density results to be identical to the temperature dependence of the normalized relaxation rate, as it should be for a $\kappa \simeq 2$ superconductor at fields $B > 0.1 \cdot B_{c2}$. Both are well represented by the clean limit BCS curve, suggesting that CaC_6 is an s-wave BCS superconductor in the clean limit regime.

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